

Orientational glass transition in C_{60} .

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We construct a model for qualitative description of the orientational glass transition in C_{60} on the spin-glass theory basis. The physical origin of the frustration and the disorder is discussed.

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As is established in a number of experiments, C_{60} crystalizes in a face centered cubic (fcc) structure. At ambient temperature the molecules rotate almost freely with centers on the fcc lattice sites, so that the space group is $Fm\bar{3}m$. When the temperature decreases to $T_c \approx 260$ K the first order orientational phase transition takes place: the sites of the initial fcc lattice become divided between four simple cubic sublattices with its own preferable molecular orientation in each sublattice. The broken symmetry group is $Pa\bar{3}$.

Moreover the neutron-diffraction experiments [see, e.g., reviews 1,2] have shown that the orientations in the ordered state are so that the electron-rich regions (the interpentagon double bonds) face the electron-deficient regions of the neighboring C_{60} molecule: the centers of pentagons or the centers of hexagons.

Solid C_{60} undergoes a well-known glass transition at $T_g = 90$ K when no orientational motion can usually be detected.^{1,2,3,4,5,6,7,8,9} Each C_{60} molecule has a single misorientation separated from the ground-state orientation by a large energy barrier E_g . The energy difference between these two orientations is U and $E_g \gg U$.^{1,3,4,10,11} These two minima of the intermolecular angle dependent energy were shown to be much lower than the energies of other mutual orientations of the pair of molecules.^{2,12,13} Below T_g molecules orientations are forming glass and the occupation probability of misoriented molecules is essentially frozen at value of $\approx 17\%$. And this static orientational disorder persists down to very low temperatures. The thermal energy is too small compared with the energy threshold between the two states for further reorientation to be possible.

In spite of the recent progress the present understanding of the intermolecular interactions in solid C_{60} is still imperfect and no single model is able to describe correctly its whole physical properties.^{12,13} This paper discusses glassy orientational transition in C_{60} . The glass state in C_{60} usually was described purely phenomenologically as “freezing” of an ensemble of two-level systems. The step towards microscopic description of glassy orientational transitions in ortho-para hydrogen was developed recently in Refs.14,15; the developed approach is useful because it takes carefully into account disorder and frustration. The symmetry of C_{60} molecules essentially differs from the symmetry of hydrogen; the method used in Refs.14,15 can not be directly used for glassy orientational transitions in C_{60} . We show how the approach of

Refs.14,15 should be modified for C_{60} case.

Now we follow the consideration of the Ref.16. We consider the restricted number of allowed orientations instead of the continuous rotations. Let us take into account in the energy only the orientations with pentagons, hexagons or double bonds directed towards 12 nearest neighbors in fcc-lattice. The C_{60} molecule is constructed in such a way that if 6 of its 12 pentagons (or 6 of its 20 hexagons) face 6 nearest neighbor double bonds (P and H states) then 6 of its 30 interpentagon double bonds face the remaining nearest neighbors. Now the energy matrix elements can take only three values; J_0 , the energy of the general mutual position, J_p pentagon versus double bond, and J_H , hexagon versus double bond. Following Ref.17 and putting $J_0 = 0$ we obtain from Fig.(2b) of Ref.10 $J_P = -300$ K and $J_H = -110$ K. The large number of the nearest neighbor bonds allows for the mean-field description of the problem.

We do not use the multipole expansion. So in the framework of our model calculations it is possible to build up the allowed functions using only the harmonics with $l = 6$. We need only their transformation properties and effectively take into account higher order terms.

Let us construct the functions $P_i(\omega)$ and $H_i(\omega)$ explicitly in terms of cubic harmonics $K_m = K_{6,m}$, $m = 1, 2, \dots, 13$ (see, e.g., Ref.18). All functions P_i and H_i are the sums of K_m , invariant under the icosahedral symmetry of the molecule (i.e., belonging to the A_{1g} representation of the icosahedral group I_h) if icosahedrons are naturally oriented in one of 8 properly chosen coordinate systems. The states $P_i(H_i)$ have 6 pentagons (hexagons) and 6 double bonds directed towards 12 nearest neighbors along different [100] axes. $P_1(\omega)$ describes the molecule rotated from the standard orientation B (following Ref.19) about [111] axis through the angle 97.76125° . The angle for $H_1(\omega)$ is 37.76125° . The functions $P_2(\omega)$, $P_3(\omega)$ and $P_4(\omega)$ (or H_2 , H_3 , H_4) are obtained from $P_1(\omega)$ (H_1) by subsequent counterclockwise rotations of the molecule by 90° around z axes.

If written in standard coordinate frame with cartesian axes along the cube sites these functions have the following explicit form:

$$P_1(\omega) = \alpha_P K_1(\omega) + \beta_P [K_8(\omega) + K_9(\omega) + K_{10}(\omega)] + \gamma_P [K_{11}(\omega) + K_{12}(\omega) + K_{13}(\omega)], \quad (1)$$

$$P_2(\omega) = \alpha_P K_1(\omega) + \beta_P [-K_8(\omega) + K_9(\omega) - K_{10}(\omega)] \\ + \gamma_P [-K_{11}(\omega) + K_{12}(\omega) - K_{13}(\omega)], \quad (2)$$

$$P_3(\omega) = \alpha_P K_1(\omega) + \beta_P [K_8(\omega) - K_9(\omega) - K_{10}(\omega)] \\ + \gamma_P [K_{11}(\omega) - K_{12}(\omega) - K_{13}(\omega)], \quad (3)$$

$$P_4(\omega) = \alpha_P K_1(\omega) + \beta_P [-K_8(\omega) - K_9(\omega) + K_{10}(\omega)] \\ + \gamma_P [-K_{11}(\omega) - K_{12}(\omega) + K_{13}(\omega)], \quad (4)$$

with $\alpha_P = -0.38866$; $\beta_P = 0.31486$; $\gamma_P = -0.42877$. The functions $H_i(\omega)$ have the same form as $P_i(\omega)$ but with the coefficients $\alpha_H = 0.46588$; $\beta_H = 0.37740$; $\gamma_H = 0.34432$. The functions are normalized to unity.

In the mean-field approximation by minimizing the orientational free energy one can obtain the nonlinear integral equation for the one-particle orientational distribution functions $g_i(\omega)$ for a molecule on i th sublattice of fcc C_{60} . In the neighborhood of the bifurcation point T_b we have a linearized system:

$$h_1(\omega) + \frac{1}{4\pi T} \int d\omega' [B(\omega, \omega') h_2(\omega') + A(\omega, \omega') h_3(\omega') \\ + D(\omega, \omega') h_4(\omega')] = 0, \quad (5)$$

where $g_i(\omega) = \mu h_i(\omega) + \dots$; $\mu = 1/T - 1/T_b$ and analogously for three other sublattices.

Here $A(\omega, \omega')$, $B(\omega, \omega')$, $D(\omega, \omega')$ are the sums of interactions over the nearest neighbors in the sublattices. For example, the sum in the plane perpendicular to the x axis can be written explicitly in the form

$$D(\omega, \omega') = 2 \{ [(P_1(\omega) + P_4(\omega)) J_P + (H_1(\omega) + H_4(\omega)) J_H] \\ \times [P_2(\omega') + P_3(\omega') + H_2(\omega') + H_3(\omega')] \\ + [P_2(\omega) + P_3(\omega) + H_2(\omega) + H_3(\omega)] \\ \times [(P_1(\omega') + P_4(\omega')) J_P + (H_1(\omega') + H_4(\omega')) J_H] \\ + [(P_2(\omega) + P_3(\omega)) J_P + (H_2(\omega) + H_3(\omega)) J_H] \\ \times [P_1(\omega') + P_4(\omega') + H_1(\omega') + H_4(\omega')] \\ + [P_1(\omega) + P_4(\omega) + H_1(\omega) + H_4(\omega)] \\ \times [(P_2(\omega') + P_3(\omega')) J_P + (H_2(\omega') + H_3(\omega')) J_H] \}. \quad (6)$$

We add the condition $h_i(\omega)$ to transform one into another under the action of the cubic group rotation elements which leave the fcc lattice invariant. At the bifurcation point $T_b > 0$ nontrivial solution with broken symmetry appears, corresponding to the orientationally ordered phase. We have $T_b = 275\text{K}$ ($T_c^{(\text{exp})} \approx 260\text{K}$) and

$$h_1(\omega) = aP_1(\omega) + bH_1(\omega) + cK_1(\omega), \quad (7)$$

$$h_2(\omega) = aP_3(\omega) + bH_3(\omega) + cK_1(\omega), \quad (8)$$

$$h_3(\omega) = aP_4(\omega) + bH_4(\omega) + cK_1(\omega), \quad (9)$$

$$h_4(\omega) = aP_2(\omega) + bH_2(\omega) + cK_1(\omega), \quad (10)$$

$$a\alpha_P + b\alpha_H + c = 0. \quad (11)$$

So we obtain¹⁶ the bifurcation temperature, the symmetry of the ordered phase and the ratio ρ of the number of molecules with pentagon facing neighbor double bond near the phase transition in good agreement with the experimental data:

$$\rho_P = \frac{a}{a+b} = 0.608, \quad (12)$$

$$\rho_H = \frac{b}{a+b} = 0.392. \quad (13)$$

The experiments show [see, e.g., Refs.1,2] that the ratio of the number of molecules in those two states is about 60 : 40 at the phase transition temperature, and increases when the temperature decreases and freezes at T_g .

It is convenient to reformulate our results. In the orientational ordered phase all sublattices are physically equivalent: they convert to each other when we rotate the crystal on 90° along the z -axis. The expressions for h_2 , h_3 and h_4 formally coincide with $h_1(\tilde{\omega})$ in their "own" system of coordinates that we label by tilde. Then we can write Eqs.(5) as a single equation as follows:

$$h_1(\tilde{\omega}) + \frac{1}{4\pi T} \int d\tilde{\omega}' E(\tilde{\omega}, \tilde{\omega}') h_1(\tilde{\omega}') = 0, \quad (14)$$

where

$$\int d\omega' [B(\omega, \omega') h_2(\omega') + A(\omega, \omega') h_3(\omega') + \\ + D(\omega, \omega') h_4(\omega')] \equiv \int d\tilde{\omega}' E(\tilde{\omega}, \tilde{\omega}') h_1(\tilde{\omega}'). \quad (15)$$

The matrix elements of E are symmetrical. They depend on J_P , J_H and the coefficients $\alpha_{P(H)}$, $\beta_{P(H)}$, $\gamma_{P(H)}$. The E-matrix can be diagonalized:

$$E(\tilde{\omega}, \tilde{\omega}') = \frac{1}{4\pi T_b} \left\{ E_r [K_8(\tilde{\omega}) + K_9(\tilde{\omega}) + K_{10}(\tilde{\omega})] + \right. \\ \left. + E_S [K_{11}(\tilde{\omega}) + K_{12}(\tilde{\omega}) + K_{13}(\tilde{\omega})] \right\} \times \\ \left\{ E_r [K_8(\tilde{\omega}') + K_9(\tilde{\omega}') + K_{10}(\tilde{\omega}')] + \right. \\ \left. + E_S [K_{11}(\tilde{\omega}') + K_{12}(\tilde{\omega}') + K_{13}(\tilde{\omega}')] \right\}. \quad (16)$$

The solution of the Eq.(14) is the following:

$$h(\tilde{\omega}) = r [K_8(\tilde{\omega}) + K_9(\tilde{\omega}) + K_{10}(\tilde{\omega})] + \\ + s [K_{11}(\tilde{\omega}) + K_{12}(\tilde{\omega}) + K_{13}(\tilde{\omega})]. \quad (17)$$

The coefficients in Eqs.(16)-(17) are determined at the bifurcation point T_b .

Equations (7)-(11) and Eq.(17) give the analytical solution for the distribution functions near the bifurcation point. It is not difficult to find the solution of the basic nonlinear integral equations for the distribution functions

in wider range of temperatures. This solution preserves the shape of Eqs.(7-11) and Eq.(17) with temperature dependent coefficients $a(T)$, $b(T)$ and $c(T)$ [$r(T)$ and $s(T)$]. But the relative ratio of the molecules number [that is determined by $a(T)/b(T)$ and $r(T)/s(T)$ ratio] weakly depend on the temperature that does not correspond to the experimental data. The two minima in the potential energy [correspond to P and H orientations] are separated by rather high barrier^{1,2,3,11}, that is not taken into account in our model, Eq.(6), so we introduce $J_P = J_P(T)$ and $J_H = J_H(T)$ to correct it.¹⁷ It is obvious that the pair interactions between molecules become less sensible to their mutual orientations when the temperature increases because the libration increases. The theoretical estimate¹⁰ shows that the libration amplitude about [111] direction increases nearly twice when the temperature changes from T_g to T_c .

We tried to fit experimental data^{1,2} to our solution, Eqs.(7)-(11). It followed that when $\rho_P \rightarrow 0.7$ from below then $T_b \rightarrow 150\text{K}$ from above and $J_H \rightarrow 0$ [J_H changes sign at this value of ρ_P , but $J_P < 0$]. Two types of mutual molecular orientations are profitable above this temperature: a) the pentagon of one molecule versus the double bond of the neighbor molecule or b) the hexagon of one molecule versus the double bond of the neighbor molecule. Below this concentration, $\rho_P = 0.7$, the frustration appears: (a)-interaction is profitable and (b)-interaction is not. Let us remind that in our model the presence of the barriers between the energy levels is taken into account indirectly: through the effective parameters J_P and J_H .

Optical experiments show that the orientational disorder at low temperatures leads to the nonhomogeneous lattice deformation, see Refs.7,8 and refs. therein. The neutron scattering data show that at low enough temperatures a relatively large fraction of the intermolecular contacts were with double bonds pointing to pentagonal or hexagonal faces. Only a small fraction of molecules were oriented at random. But this picture never led to the low temperature phase, but only to partly frustrated structures.²² It follows from the experimental data^{1,7} that there is small anomaly at $T \approx 150\text{K}$ but it is still unclear whether this temperature corresponds to the arrest of the free uniaxial rotation or to a glass transition similar to that observed near 90K. Note that the discrete description of the orientational ordering, Eqs.(7)-(11), includes as the answer the equiprobable distribution of the molecule orientations in the cubic lattice, $K_1(\omega)$, that corresponds to the rotations in cubic lattice [see, e.g., Ref.20] in addition to H and P states.

In summary at high temperatures the molecules travel slightly between different types of mutual orientations. When the temperature decreases the possibility to change the orientation for the molecule decreases because of large potential barrier between the lowest orientational states. Then the H -state of the molecule is rather rare phenomenon at low temperatures. The glassy state is obtained when the thermal energy is not sufficient to over-

come the potential barrier that separates the two orientational configurations^{1,2}. The H -state of the molecule [6 hexagons and 6 double bonds] is then profitable from the double bond side and is not from the hexagon side. The P -state of the molecule [6 pentagons and 6 double bonds] is then profitable from the double bond side and from the pentagon side. We see that the behavior of our system at low temperatures is similar to the behavior of the “dilute” molecular crystals, e.g. ortho-para hydrogen.^{14,15} H -states here play the role of $p - H_2$ molecules in ortho-para mixture.

The solution, Eq.(17) [and Eqs.(7-11)], correctly describes symmetrically oriented order at all temperatures. But the coefficients r and s (a , b and c) should be determined from the experimental data for ρ_P and ρ_H . On this basis we suggest the following model for the glass description in C_{60} .

Let us consider a system of particles on lattice sites i, j with Hamiltonian

$$H = -\frac{1}{2} \sum_{i \neq j} J_{ij} \hat{U}_i \hat{U}_j, \quad (18)$$

where J_{ij} are quenched Gaussian interactions with zero mean,

$$P(J_{ij}) = \frac{1}{\sqrt{2\pi}J} \exp \left[-\frac{(J_{ij})^2}{2J^2} \right], \quad (19)$$

with $J = \tilde{J}/\sqrt{N}$.

$$\begin{aligned} \hat{U} = & c [K_8^i(\tilde{\omega}) + K_9^i(\tilde{\omega}) + K_{10}^i(\tilde{\omega})] \\ & + d [K_{11}^i(\tilde{\omega}) + K_{12}^i(\tilde{\omega}) + K_{13}^i(\tilde{\omega})], \end{aligned} \quad (20)$$

where c and d depend on the ratio of the P - and H -states [$d/c = -0.914$ when $\rho_P = 0.83$, i.e. at the pressure $P = 0$, $T = T_g$].

Using replica approach we can write the free energy averaged over disorder in the form:

$$\begin{aligned} \langle F \rangle_J / NkT = & \lim_{n \rightarrow 0} \frac{1}{n} \max \left\{ \frac{t^2}{4} \sum_{\alpha} (p^{\alpha})^2 + \frac{t^2}{2} \sum_{\alpha > \beta} (q^{\alpha\beta})^2 - \right. \\ & \left. - \ln \text{Tr}_{\{U^{\alpha}\}} \exp \left[\frac{t^2}{2} \sum_{\alpha} p^{\alpha} (\hat{U}^{\alpha})^2 + t^2 \sum_{\alpha > \beta} q^{\alpha\beta} \hat{U}^{\alpha} \hat{U}^{\beta} \right] \right\}. \end{aligned} \quad (21)$$

Here $t = \tilde{J}/kT$, $\text{Tr}(\dots) \equiv \int_0^{2\pi} d\varphi \int_0^{\pi} d\cos(\theta) (\dots)$.

The saddle point conditions for the free energy give the glass and regular order parameters

$$q^{\alpha\beta} = \frac{\text{Tr} [\hat{U}^{\alpha} \hat{U}^{\beta} \exp(\hat{\theta})]}{\text{Tr} [\exp(\hat{\theta})]}, \quad (22)$$

$$m^{\alpha} = \frac{\text{Tr} [\hat{U}^{\alpha} \exp(\hat{\theta})]}{\text{Tr} [\exp(\hat{\theta})]}, \quad (23)$$

and the auxiliary order parameter

$$p^\alpha = \frac{\text{Tr}[(\hat{U}^\alpha)^2 \exp(\hat{\theta})]}{\text{Tr}[\exp(\hat{\theta})]}. \quad (24)$$

Here

$$\hat{\theta} = \frac{t^2}{2} \sum_\alpha p^\alpha (\hat{U}^\alpha)^2 + t^2 \sum_{\alpha > \beta} q^{\alpha\beta} \hat{U}^\alpha \hat{U}^\beta. \quad (25)$$

In the replica symmetric (RS) approximation²³ the free energy (21) has the form:

$$F = -NkT \left\{ t^2 \frac{q^2}{4} - t^2 \frac{p^2}{4} + \int_{-\infty}^{\infty} \frac{dz}{\sqrt{2\pi}} \exp\left(-\frac{z^2}{2}\right) \ln \text{Tr}[\exp(\hat{\theta})] \right\}. \quad (26)$$

Here

$$\hat{\theta} = zt\sqrt{q}\hat{U} + t^2 \frac{p-q}{2} \hat{U}^2. \quad (27)$$

The extremum conditions for the free energy (26) give the following equations for the glass and regular order parameters:

$$q = \int dz^G \left\{ \frac{\text{Tr}[\hat{U} \exp(\hat{\theta})]}{\text{Tr}[\exp(\hat{\theta})]} \right\}^2, \quad (28)$$

$$m = \int dz^G \left\{ \frac{\text{Tr}[\hat{U} \exp(\hat{\theta})]}{\text{Tr}[\exp(\hat{\theta})]} \right\}, \quad (29)$$

and the auxiliary equation

$$p = \int dz^G \frac{\text{Tr}[\hat{U}^2 \exp(\hat{\theta})]}{\text{Tr}[\exp(\hat{\theta})]} \quad (30)$$

Here

$$\int dz^G = \int_{-\infty}^{\infty} \frac{dz}{\sqrt{2\pi}} \exp\left(-\frac{z^2}{2}\right). \quad (31)$$

The replica symmetric solution is stable unless the replicon mode energy λ is nonzero. For our model we have:

$$\lambda_{\text{RS}} = 1 - t^2 \times \int dz^G \left\{ \frac{\text{Tr}[\hat{U}^2 \exp(\hat{\theta})]}{\text{Tr}[\exp(\hat{\theta})]} - \left[\frac{\text{Tr}[\hat{U} \exp(\hat{\theta})]}{\text{Tr}[\exp(\hat{\theta})]} \right]^2 \right\}. \quad (32)$$

The results of the calculations are illustrated in Fig.1. The order parameters do not go to zero because

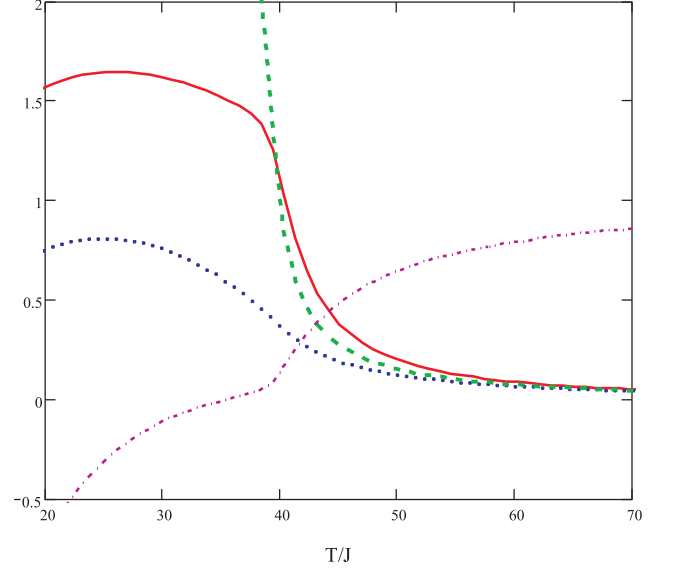


FIG. 1: Order parameters and the heat capacity evolution with the temperature. Here $d/c = -0.914$, red solid curve is the heat capacity, dotted blue and dash green curves are the order parameters m and \sqrt{q} respectively, violet dashed curve is the replicon mode λ_{repl} . The replica symmetry breaking occurs at the temperature T_0 corresponding to $\lambda_{\text{repl}} = 0$; the glass transition temperature T_g usually is very close to T_0 .

$\int d\omega u^{2k+1}(\omega) \neq 0, k = 0, 1, \dots$ ²⁴ The co-existence of the orientational ordered state (the order parameter m) and the glass (the order parameter q) is in agreement with the experimental data.^{1,2,4,5,6}

The replica symmetry breaking occurs at the temperature T_0 corresponding to $\lambda_{\text{repl}} = 0$; the glass transition temperature, T_g , usually is very close to T_0 . The mildly sloping curve with the broad maximum for the heat capacity qualitatively corresponds to the curve $C_p^{\text{exp}}(T)$ obtained in Ref.3.

It is well known that the temperature of orientational transition increases with pressure.^{1,2} When the pressure increases the number of P -states decreases, but the number of H -states rises. It is known that $J_P = J_H = J$ and $\rho_H = 0.5$ when the pressure $P \approx 0.25$ Gpa and $T \approx 300$ K [see Fig.13 in Ref.1]. Then we get from our model using the bifurcation condition: $J = -242$ K. If we assume that J_P and J_H depend linearly on P then we get:

$$J_P = -300(\text{K}) + 230(\text{K/Gpa})P, \quad (33)$$

$$J_H = -110(\text{K}) - 530(\text{K/Gpa})P. \quad (34)$$

Then if $P = 0.1$ Gpa then we find $T_c = 281$ K and $\rho_P = 0.56$; if $P = 0.2$ Gpa then we find $T_c = 293$ K and $\rho_P = 0.52$; if $P = 0.3$ Gpa then we find $T_c = 309$ K and $\rho_P = 0.48$. These results agree well with the experimental data¹ for T_c and ρ_P . We believe that our approach describes well the orientational transition at small pressure.

When $P \rightarrow 1.3\text{GPa}$ it follows from Eqs.(33)-(34) that $J_P \rightarrow 0$ [it changes its sign here] and $J_H < 0$. So our simple model for the orientational ordering becomes invalid. Experiments show^{1,2,4,5,6} that at these pressures the number of P -states is very small.

The orientational glass transition is hardly experimentally seen at $P \sim 0.2\text{GPa}$ [$\rho_P \approx \rho_H$ at all temperatures¹] and $P \gtrsim 1.5\text{GPa}$ [$\rho_P \ll 1$] that does not contradict the above description of the glass transition. When $\rho_P \approx \rho_H$ the P - and H -states are both profitable and the analogy

with diluted multipole systems becomes invalid. When the pressure is large enough ρ_P becomes very small and there is no sense speaking about disorder and frustration.

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